A method for measuring the high temperature emissivity of refractory metal surfaces

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Abstract

For high temperature applications, such as lighting components or heating elements, the thermal emissivity of refractory metals such as tungsten can be improved by altering the surface of the material. Laser structuring or porous sintered coatings have been shown to increase the emissivity, which leads to improvements in process stability and component lifetime.

Measuring the emissivity is generally performed at room temperature by reflectometry as high temperature measurement is extremely difficult. This is a significant disadvantage for comparing surface modifications, as emissivity at application temperature might differ greatly. In this paper, a method is described with which samples are heated by a direct current flow and the emissivity is measured with an optical pyrometer by comparing temperature values with the actual surface temperature.

The method allows a comparative measurement of different surface treatments at the application temperature. Degradation of the surface emissivity at the application temperature is also possible to observe. Measured values for tungsten with different surface modifications are shown and discussed.

Keywords

refractory metals, lighting components, heating element, high temperature emissivity, surface treatment

Introduction

For high temperature engineering applications, the emissivity is an important property. An increased emissivity is often highly favourable to ensure sufficient cooling, to increase the energy efficiency or to provide stable and homogeneous process conditions. This provides benefits in very different fields of research, including the obvious thermal processes in power plants and furnaces [1–3]. In other areas, such as the aeronautical and spacecraft industry [4–8] or the lighting industry [9, 10], heat is often generated as an undesired byproduct. In these applications, surfaces with increased emissivity are used to remove the heat from the system to ensure longer component life time.

As the emissivity is strongly dependent on the surface conditions, surface structure and material composition are both important factors for optical parameters. The relationship between structure and emissivity has been studied in experimental investigations [1, 11–14] as well as modelling approaches [2, 3, 15, 16].

When radiation interacts with a sample, it may partially be reflected (reflection ρ), absorbed (absorption α) or transmitted (transmission τ). These characteristics are tied together by the principle of energy conservation as shown in equation (1).

$$1 = \propto + \rho + \tau \quad (1)$$

According to Kirchhoff's law of thermal radiation, the emissivity ε of an opaque body (τ=0) is equal to its absorption α at a given wavelength λ and temperature T. This can approximately be applied to obtain the emissivity via equation (2) where ρ is the reflectivity and β, φ specify the observing angle [17].

$$\varepsilon(\lambda, \mathrm{T}, \beta, \varphi) = \alpha(\lambda, \mathrm{T}, \beta, \varphi) = 1 - \rho(\lambda, \mathrm{T}, \beta, \varphi) \quad (2)$$

The emissivity ε is defined as the ratio of energy radiated by the sample to an ideal thermal emitter, a blackbody. Therefore, real objects in use show an emissivity of ε<1. While this method has its limitations because of non-ideal diffusely reflecting surfaces, it is often accurate enough to compare similar opaque bodies.

Because of the temperature dependency of ε, it would be of interest for high temperature applications (>1600 °C), to measure different surface modifications in-situ. Since this is almost impossible to achieve, a method will be discussed in this paper to compare various surface conditions close to application temperature. For this, tungsten samples with different surface modifications were heated by a direct current flow up to 1800 °C. The emissivity was then determined by comparing the optically measured surface temperature on the modified area of the sample with the actual surface temperature. The emissivity value stored in the pyrometer used for the optical measurement can then be altered until the two readings are equal. The ε value for which this condition is met, is the emissivity of the surface. Several methods to obtain the actual surface temperature were used and are described in detail below.

Experimental

Experimental setup

When measuring the high temperature emissivity, special care has to be taken of how the heat is being generated and transferred to the sample surface. Simply heating the sample by thermal radiation from heating elements is not viable, because reflections of the heat emitted from the external heating elements interfere with the measurement on the sample surface. Furthermore, the sample surface needs to be accessible by optical measurement using a pyrometer. Because of the formation of oxides in air, the measurement has to be performed under inert atmosphere. Moreover in the case of tungsten, volatile oxides are formed under oxidizing conditions. By design, this excludes most available furnaces in which the sample of interest can be heated up to 1800 °C.

Argon atmosphere, direct current flow and optical accessibility were all found in a sintering press available at Plansee SE in Reutte. A FAST/SPS sintering press (DSP 515 from Dr. Fritsch, as shown in

Fig. 1) was adapted for emissivity measurements as shown in Fig. 2. The sintering press was operated without powder, the ring mould and punches, but instead by using a W rod as the sample to be heated. A graphite foil was used on top and on the bottom of the sample to minimize gaps between the sample and the electrodes providing better electrical contact. As the sintering press has to be operated at a minimal load of 28 kN, the tungsten sample was designed to be 40 mm in diameter to withstand plastic deformation at 1800 °C.

As unwanted oxidation is one of the main difficulties to overcome for measuring high temperature emissivity, metallic foils were added to the setup to ensure minimal oxygen uptake by the sample by reducing oxygen access (Mo foil) or by acting as an O_2-getter (Ta-foil). A graphite mat and a ring-shaped graphite casing to suppress reflections and further reduce oxygen access completed the experimental setup. Optical access to the sample was made possible by a window in the graphite casing. A picture of the sample inside the graphite casing is shown in Fig. 3. To exclude carburization of the tungsten surface, a test run with a pure W sample was performed. X-ray diffraction phase analysis did not show any sign of carburization after a complete run up to 1800 °C.

Figure 1: FAST/SPS sintering press DSP 515 from Dr. Fritsch used for the measurements.

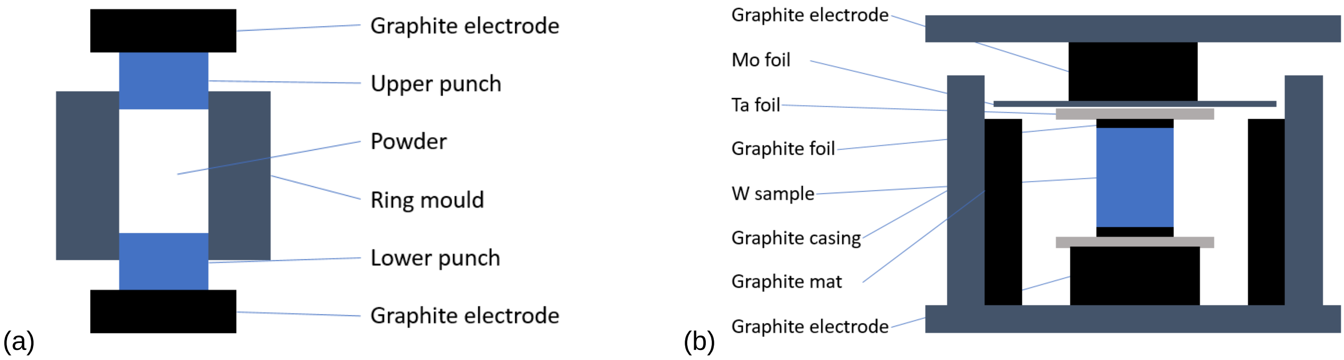

Figure 2: (a) Original setup for direct current sintering. (b) Adapted experimental setup for high temperature emissivity measurements.

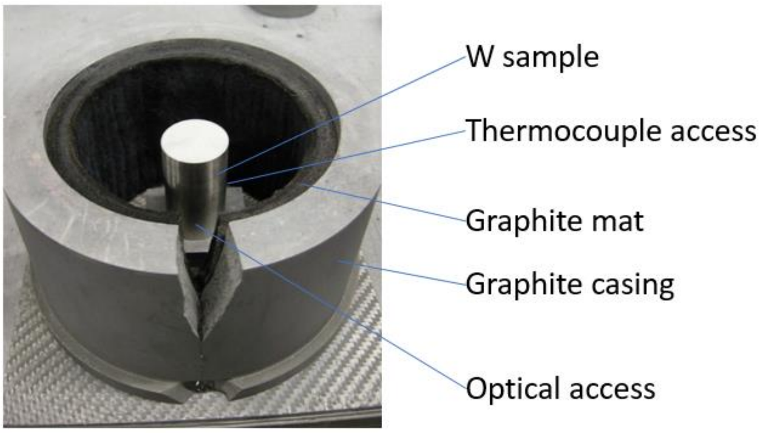

Figure 3: W sample inside the graphite casing and mat.

Another challenge for investigating high temperature emissivity is measuring the actual surface temperature of the sample. Two separate but both accurate temperature measurements are necessary for comparison. In principle, there are two ways to determine the sample temperature, by using a thermocouple or optically by pyrometric measurement. Both alternatives have been tested and shall be discussed in detail.

Temperature measurement

A very common method for temperature measurement is using a thermocouple. This allows for simultaneous measurement of the surface temperature and comparison to the optical signal from the pyrometer on the area of interest. The most significant advantage over optical measurements is that there are no assumptions on the material properties of the sample - such as the emissivity - needed to interpret the signal. On the other hand, measuring surface temperatures of up to 1800 °C by thermocouple is no straightforward task.

W-Re thermocouples which are suited to measure such high temperatures are very stiff and therefore difficult to bring into contact with the sample surface. Since it neither would be feasible to move a welded thermocouple together with the sample to the sintering press, nor can the connection be made in-situ, physical contact with the sample surface had to be established by contacting the thermocouple to the sample mechanically. The thermocouple was held in place with an alumina rod which was fastened to the sample with molydenum wire. This had to be done in such a way as to ensure that the connection of thermocouple wires was as close to the sample surface as possible. On top of that, the thermocouple had to be placed at the same vertical position as the pyrometer, due to a vertical temperature gradient observed on the sample. Fig. 4 illustrates the temperature measurement by thermocouple as performed for the tests presented in this paper.

In addition to bad contact of the thermocouple with the sample surface and the temperature gradient of the sample, the inherent mass of the W-Re wires and therefore their cooling effect were the main sources of error observed throughout the measurements.

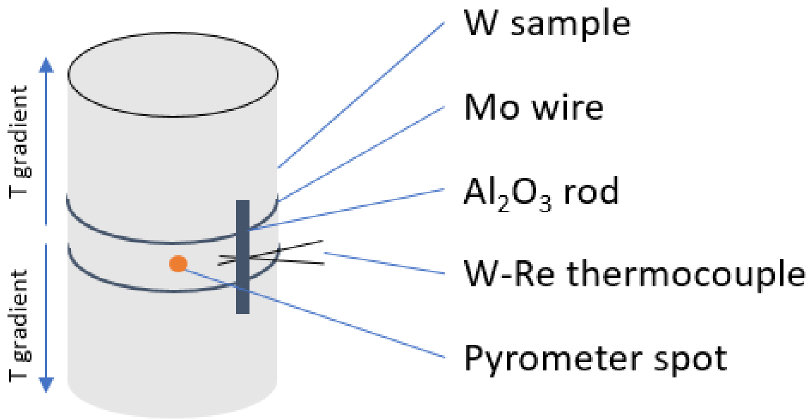

Figure 4: Temperature measurement by thermocouple.

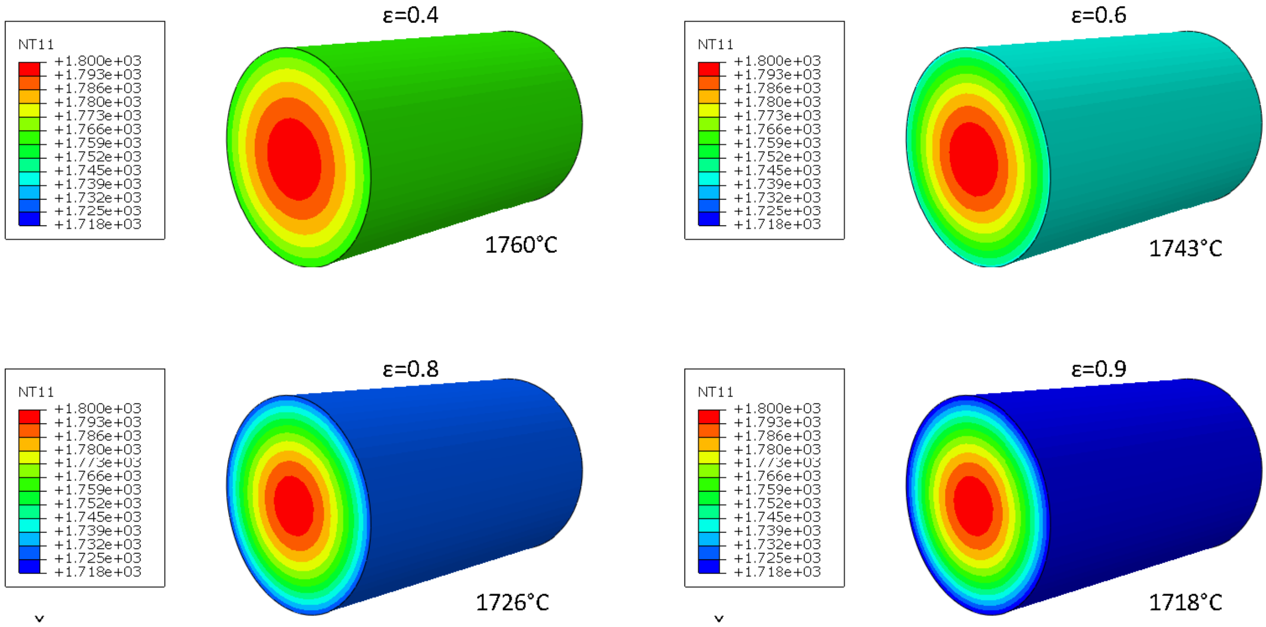

Figure 5: Calculated surface temperature for various surface emissivities ε. Core temperature was set to 1800°C in all cases.

Another method of temperature determination by thermocouple would be to measure the core temperature and estimate the surface temperature from that. While this would seem to eliminate the aforementioned sources of error, the surface temperature depends on the emissivity of the surface itself. The influence of the surface emissivity on the surface temperature is illustrated in Fig. 5. For these calculations, the core temperature of a tungsten rod with a diameter of 40 mm was fixed at 1800 °C with surface emissivities ranging from ε=0.4 to ε=0.9. Calculated surface temperatures range from 1760 °C to 1718 °C. In conclusion, measuring the core temperature for estimating the surface temperature of the sample requires good knowledge of the surface emissivity in advance. From Fig. 5, it is also obvious how important correctly determining the surface temperature for emissivity measurements is.

Instead of measuring the surface temperature by thermocouple, it can also be determined optically by pyrometric measurement. Pyrometers in use are calibrated against a cavity simulating an ideal blackbody. In order to serve as a blackbody, the cavity needs to fulfill certain conditions [18]. First and foremost, it needs to be wider than the spot size of the pyrometer. Secondly, the ratio between depth and width should be in the range of 10–12, depending on the emissivity of the material inside the cavity. As a cavity like this cannot be easily drilled into tungsten, mechanical preparation on every sample would have been very expensive and was therefore avoided. Another difficulty when using this method is to compensate for thermal expansion of the sample. Because of that, the position of the cavity cannot easily be set at room temperature. On the other hand, at 1800 °C, the sample including the cavity are so bright, visual adjustments by laser optics are not viable. To compare the results to our high temperature thermocouple measurements, this method to determine the surface temperature was performed on one sample. The emissivity of the tungsten surface in "as ground" condition was found to be 0.40 compared to 0.39 as observed by thermocouple temperature determination.

For future measurements, temperature determination by 2-colour pyrometry would be the preferred choice. Here, the temperature is determined by optical measurement using two different wavelengths simultaneously. Under the assumption that the surface shows the same emissivity for both wavelengths, this measurement is independent of the absolute value of surface emissivity. After determining the surface temperature by 2-colour pyrometry, an ordinary single-colour pyrometer could then be aimed at the sample. As when using any other method of temperature determination, the emissivity value stored in the pyrometer can then be altered until the two temperature readings are equal.

Samples for emissivity measurements

Tungsten rods with a diameter of 40 mm and a length of 60 mm served as samples for the measurements. By mechanical preparation, one side of the cylinder was prepared flat in a perpendicular position to the bottom and top surfaces. For better understanding, a sketch of the sample in top-view is provided and set into relation of the optical measurement in Fig. 6. This flat surface was then modified by laser structuring or by applying a coating. Defined troughs in periodic sequence in a sawtooth-like pattern were manufactured by laser ablation. Coatings were applied via suspension technique, where a powder is deposited on the surface at room temperature and then sintered to the surface. Through this process, a porous sintered coating which can offer the benefits of both increased surface area as well as the ability to alter the chemical composition of the surface is possible. The different surface modifications tested for this work are listed in Table 1. All samples were heated to and held at 1900 °C for 1 h prior to all measurements to avoid any influence of sintering processes.

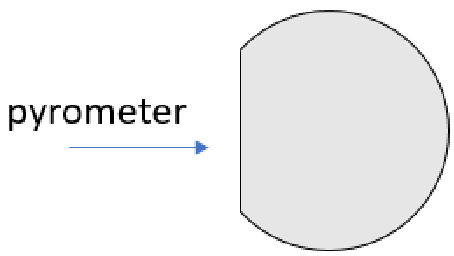

Figure 6: Tungsten sample in top-view.

Table 1: List of tested surface conditions.

Surface condition #	Type of surface
1	Pure W – as ground
2	W porous sintered coating
3	Slurry coating 1
4	Slurry coating 2
6	Laser structure 1
7	Laser structure 2

Analysis

Tungsten samples with various surface modifications were heated by a direct current flow to 1400 °C, 1600 °C and 1800 °C, cooled down to 1600 °C and 1400 °C and heated up to 1800 °C again. At every temperature step, emissivity determination was performed. For that, two separate temperature measurements by pyrometer and by thermocouple as shown in Fig. 4 were compared. The emissivity value stored in the pyrometer used for the optical measurement was altered until the two readings were equal. The ε value for which this condition is met, is the emissivity of the surface.

For the optical measurements, a SensorTherm GmbH Metis MS09 pyrometer with a wavelength range of 0.7–1.1 µm was used. SensorTherm Software was used for the interpretation of the pyrometric measurement.

High temperature emissivity values were compared to emissivity values obtained at room temperature. These measurements were performed using a 410-Solar Reflectometer by Surface Optics Corporation. From the measured directional hemispherical reflectance ρ, the diffusely reflected fraction in the wavelength range of 1000–1700 nm was used to calculate the emissivity ε of the sample according to equation (2).

Results and Discussion

In Fig. 7, high temperature emissivity values for different surface modifications obtained after first reaching the respective temperature are shown. Further measurements at the same temperature led to the same results and are therefore not displayed in the diagram.

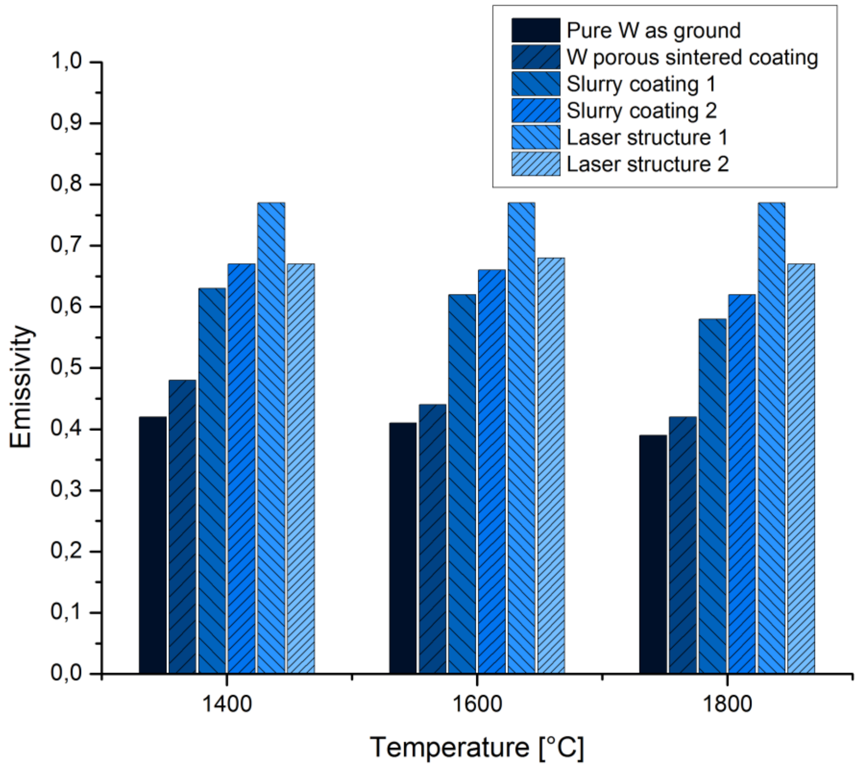

Figure 7: High temperature emissivity at 1400 °C, 1600 °C and 1800 °C for various modified surfaces.

Both coatings and laser structuring have the potential of increasing the thermal emissivity of tungsten parts. For the tested slurry coatings, the emissivity slightly decreases with increasing temperature. At very high temperatures, the W porous sintered coating shows only little benefit over the pure W surface in as ground condition. Other slurry coatings increased the thermal emissivity by a factor of 1.5 compared to the “as ground” surface.

Even higher emissivities than for slurry coatings were observed for surface structures manufactured by laser ablation. However, it is important to notice that these structures are very expensive to implement. Even for small parts such as the samples used for this work, costs of laser structuring can easily be higher by a factor of 100 over a slurry coating.

Apart from accurately determining the surface temperature, the main source of error of measurement was oxidation and therefore alteration of the samples. Some even showed discolorations after the high temperature treatment, giving proof of residual oxygen inside the sintering press. Especially the slurry coated surfaces showed signs of oxidation. To estimate the influence of oxidation on the measured values, the samples were analyzed for room temperature emissivity before and after the high temperature treatment. As can be seen in Fig. 8., the slurry coated samples show higher values before the oxidation than after due to the nature of oxides being formed. This is not the case for pure W surfaces as seen with the “as ground”-sample and the laser structured surfaces. It seems therefore that for slurry coated surfaces, the actual high temperature emissivity would be higher without oxidation than as shown in Fig. 7.

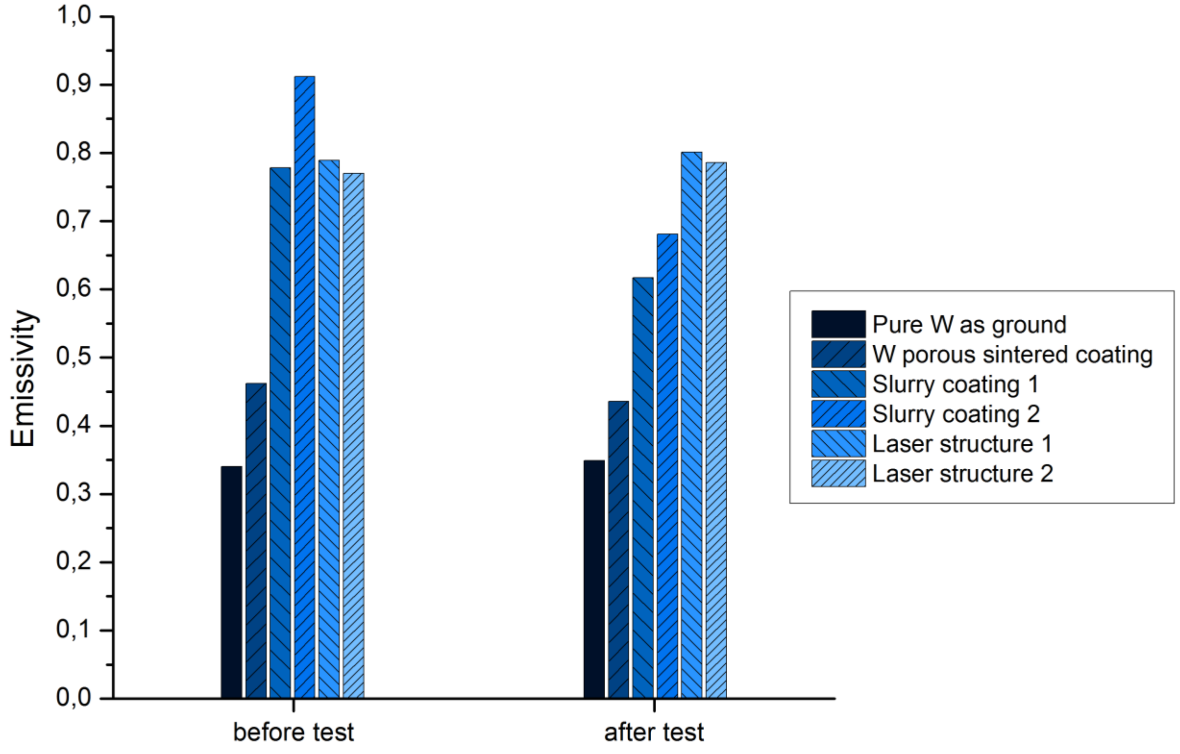

Figure 8: Spectral hemispherical emissivity at room temperature (1000-1700 nm wavelength range) before and after high temperature treatment.

For future measurements, residual oxygen needs to be removed from the chamber to avoid oxidation of the samples. Furthermore, accurate temperature determination by thermocouple has proven to be difficult. Connecting the W-Re wires to the surface had to be repeated several times for each sample until sufficient contact was reliably established. Preparing the experimental setup was therefore a very time consuming process. Using a 2-colour pyrometer and a single-colour pyrometer simultaneously should lead to a lower error of measurement in the future.

Summary

High temperature emissivity measurements were performed on tungsten samples with various surface modifications. Heating the samples by a direct current flow proves to be a feasible method to generate the necessary surface temperatures. The two main challenges for high temperature emissivity measurements were oxidation by residual oxygen inside the chamber and accurately detecting the surface temperature. Both can be overcome by adapting the experimental setup as discussed above for future trials. It could be shown that both, applying a slurry coating and laser structuring of the surface, are viable methods to improve the high temperature emissivity. When comparing the results obtained by high temperature measurements to room temperature measurements, the same trends are observed for the tested surface modifications. Concluding, developments to increase the thermal emissivity on surfaces can be monitored with comparably low effort by reflectometry at room temperature, while the most promising surface modifications can be reviewed at application temperature by the method presented in this paper.

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